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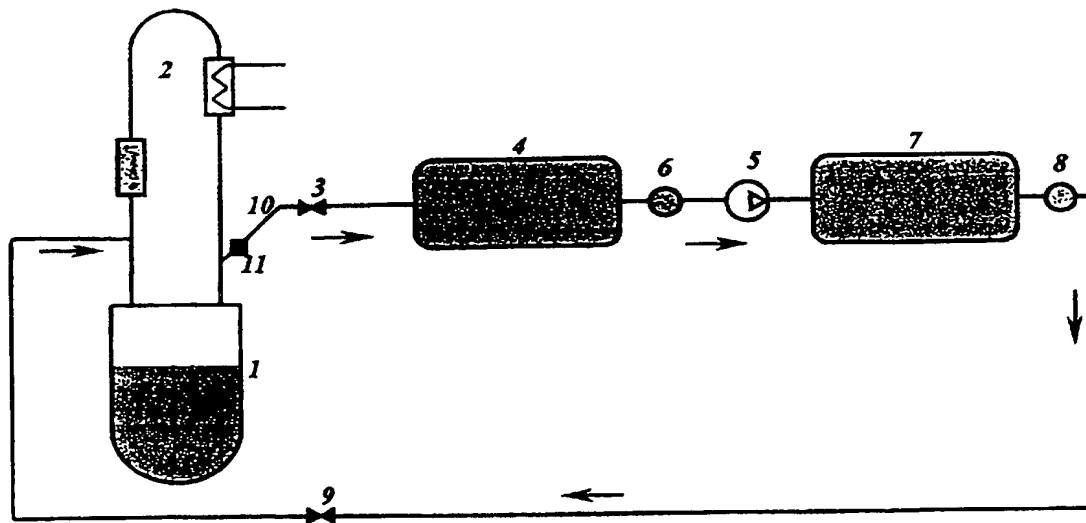
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(54) Title: METHOD OF STRONTIUM-89 RADIOISOTOPE PRODUCTION



(57) Abstract: Inert gaseous fission products, including beryllium, rubidium, and krypton isotopes, resulting from the operation of a uranyl sulfate water solution nuclear reactor (1) are passed through a delaying device (4) to precipitate out strontium-90, then passed to a second delaying device (7) to precipitate out the desired strontium-89.

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## METHOD OF STRONTIUM-89 RADIOISOTOPE PRODUCTION

### Technical Field

#### 1. Field of the Invention

5                   The present invention is in the field of radioisotope production and in particular relates to a method of producing strontium-89.

### Background Art

#### 2. Description of the Prior Art

10                   Radioisotopes have been used in nuclear medicine for diagnostics and therapeutics for more than fifty years. Medical radioisotope production is an important industry using more than 50% of the radioisotopes produced in the world. More than 160 radioisotopes of 80 chemical elements are produced with the help of nuclear reactors and  
15                   charged particle accelerators today.

One of the most effective modern therapeutic radioisotopes  
                  is strontium-89. It is used for pain palliation instead of drugs when  
                  treating cancer. When medicine containing strontium-89 is introduced  
                  into an organism, it is absorbed and distributed in the bone metastases  
20                   providing for a long anesthetic effect.

                  Strontium-89 radioisotope has a half-life of 52.7 days with  $\beta$  decay (decays to  $Y^{89}$ , a stable isotope). The maximum energy of the  $\beta$ -

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particles is 1463 keV. The attendant  $\gamma$ -radiation energy is 909.1 keV.

Strontium is a biochemical analog of calcium that has the same transport mechanism in the human body. Strontium chloride  $\text{SrCl}_2$  introduced to the vein is mainly accumulated in bone metastases providing for a long anesthetic effect so it is not necessary to take drugs frequently and the patient does not become tolerant of them. Malignant tumors tending to metastases in the skeleton are: mammary gland, large intestine, thyroid gland, prostate, kidney, and skin cancer. The maximum range of  $\beta$ -particles of strontium-89 in the bone does not exceed 7 mm, so its radiation effects are isolated to the small area of the skeleton and its radiation burden on the marrow and nearby soft tissue is not significant. As strontium-89 is incorporated in the mineral structure of the bone, diseased metabolism does not take place, and it remains there for more than 100 days. Healthy bone contains a small component of the injected dose and loses it quickly during the first fortnight. One injection of strontium chloride is about 4 mCi and is effective for 3 to 6 months. Clinical tests of the preparation based on  $^{89}\text{SrCl}_2$  showed that 65-76% of the patients said that pain had been reduced significantly, and there was full anaesthetic effect in 20% of the cases. In addition, doctors think that strontium-89 chloride has a therapeutic effect, which means it does not only block metastases but also reduces them.

One reactor method of strontium-89 production consists of

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irradiating a target of strontium carbonate  $\text{SrCO}_3$  with neutrons having a thermal neutron spectrum. A target made from metallic strontium is irradiated by the neutron flux of a nuclear reactor. Natural strontium consists of the following isotopes:  $\text{Sr}^{84}$  at 0.56%,  $\text{Sr}^{86}$  at 9.9%,  $\text{Sr}^{87}$  at 7.0% and  $\text{Sr}^{88}$  at 82.6%. The strontium-89 radioisotope is formed in the target as a result of the neutron capture reaction of one of the strontium isotopes  $\text{Sr}^{88} (n, \gamma) \text{Sr}^{89}$ . A highly enriched target containing  $\text{Sr}^{88} > 99.9\%$  is used because it is necessary to eliminate strontium-85 from the reaction  $\text{Sr}^{84} (n, \gamma) \text{Sr}^{85}$ , an undesirable admixture. This is a convenient production method and takes place in a normal research reactor. The cross-section of the  $(n, \gamma)$ -reaction is only  $6 \times 10^{-27} \text{ cm}^2$ , however, which restricts the productivity of this method.

Another strontium-89 production method is based the threshold reaction of neutron capture with the emission of a charged particle  $\text{Y}^{89} (n, p) \text{Sr}^{89}$ . A target containing natural monoisotope Yttrium-89 is irradiated in the neutron flux of a nuclear reactor with a fast neutron spectrum and is subsequently subjected to radiochemical reprocessing for extraction. Strontium-89 production can achieve about 10-15 mCi per gram of yttrium in optimum conditions. The target is a pellet of yttrium oxide  $\text{Y}_2\text{O}_3$  of high purity that is pressed and annealed at  $1600^\circ \text{ C}$ . This method produces almost no radioactive wastes and the end-product does not contain harmful admixtures, e.g., the quantity of attendant strontium-

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90 is less than  $2 \times 10^{-4}$  atomic percent.

This method has an extremely low productivity due to the small cross-section of the (n,p)-reaction on  $Y^{89}$ , less than  $0.3 \times 10^{-27} \text{ cm}^2$  for neutrons of the fission spectrum. It can only occur in reactors with a fast neutron spectrum, and there are few in existence. In addition, yttrium purified without admixtures of uranium should be used (the uranium content in the  $Y_2O_3$  pellets must not exceed  $10^{-5}$  by mass). Low productivity and the need for reactors with a fast neutron spectrum are the main problems with this method.

There is clearly a need for a more efficient method for the production of strontium-89, particularly one that uses a relatively low power reactor.

#### Disclosure of the Invention

A solution nuclear reactor containing a uranyl sulfate fuel solution produces krypton-89 during operation. Krypton-89 is in the form of a gas that bubbles to the surface of the fuel solution and occupies the enclosed volume above the fuel. An inert gas transports the krypton-89, along with other radioisotope fragments, in a sealed system to a trap area where any accompanying relatively short half-life krypton-90 is allowed to decay to strontium-90. The strontium-90 is removed. Then the krypton-89 is transported to a catching system where it remains until it fully decays to strontium-89. The strontium-89 is removed from the inert gas

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with the help of sorption in a carbon trap or by chemical interaction in an acid environment. The inert gas is returned to the reactor core.

#### Brief Description of the Drawings

- 5                      Fig. 1A            shows the fission products decay of  $\text{Br}^{89}$  and  $\text{Br}^{90}$ .
- Fig. 1B            shows the fission products decay of  $\text{Kr}^{91}$ .
- Fig. 1C            shows fission products decay of  $\text{Kr}^{92}$  and  $\text{Kr}^{93}$ .
- Fig. 2            is a schematic of the gas loop for  $\text{Sr}^{89}$  production.

10                      Description of Preferred Embodiments

                         The strontium-89 production method is based upon a unique ability to effect not only the final radioisotopes, but also its precursors produced as a result of the nuclear transformation of products in the decay chain of elements with mass 89 occurring in a nuclear solution reactor. The decay chain is  $\text{Se}^{89} \rightarrow \text{Br}^{89} \rightarrow \text{Kr}^{89} \rightarrow \text{Rb}^{89} \rightarrow \text{Sr}^{89}$ .

15

                         A liquid fuel nuclear reactor having a uranyl sulfate water solution ( $\text{UO}_2\text{SO}_4$ ) core is used in the present invention. Uranium-235 and/or uranium-233 can be used as fissionable material in the fuel solution of uranyl sulfate. The Russian Argus reactor was the particular reactor used. It used 90% enriched  $\text{U}^{235}$  in a concentration of 73.2 g/l in the water solution. The uranyl sulfate water solution volume (pH=1) was

20

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22 liters. It can be brought up to its rated power of 20 kW in 20 minutes. The thermal neutron flux density in the central channel is  $5 \times 10^{11}$  neutrons/cm<sup>2</sup>s.

Homogenous solution fuel reactors have a number of advantages over hard fuel reactors. They have large negative temperature and power reactivity effects, which provides for their high nuclear safety. The core design is much simpler. There are no fuel element cladding spacers and other parts reducing the neutron characteristics. Solution preparation is much cheaper than fuel element production. Solution fuel loading (pouring) is much easier too, and makes it possible to change the fissionable material concentration in fuel or solution volume if necessary. There can be no local over-heating provoked by power density field deformations in the core of the solution reactor, thanks to good conditions for heat transfer. These reactors are simple and reliable in operation and do not require a large staff for their operation.

A number of radioactive inert gases are produced in uranyl sulfate solution reactor during its operation, including the desired krypton-89. The majority of these gases leave the solution in the gas phase, accumulating above the liquid surface. The process by which this takes place is based on "radiolytic boiling." Gas bubbles containing water vapor and hydrogen form in the tracks of fission fragments. The vapor is

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condensed within about  $10^{-8}$  seconds and a gas bubble forms having a radius of about  $10^{-5}$  cm. Fission fragments either get into the gaseous bubble during its generation or afterwards by diffusing from the solution. They then migrate to the surface of the fuel solution. The radiolytic gas bubbles rise to the surface in only a couple of seconds, making it possible to remove relatively short-life radioisotopes, such as krypton-89. Bubbling the fuel with an inert gas can speed up this process of removal of fragment gases. Krypton-89, along with small quantities of other fission fragment elements are produced at the same time.

10                   The main chains of fission products' decay resulting in strontium radionuclides whose gaseous precursors have a half-life of more than one second are shown in FIG. 1A to 1C. One of the fission products is krypton-89 ( $\text{Kr}^{89}$ ), a radioactive isotope of the inert gas, krypton, preceding strontium-89 in the decay chain of fission products

15                   with an atomic mass of 89. It has a half-life of 3.2 minutes, decaying to rubidium-89. Rubidium-89 decays with a half-life of 15.4 minutes to the desired strontium-89. Other isotopes of krypton, however, also bubble to the surface, including the highly undesirable precursor to strontium-90, krypton-90. Krypton-90 decays in 33 seconds to rubidium-90 and in 2.91

20                   minutes to strontium-90. Because krypton-89 and krypton-90 are gases and because of the differential in half-life of the two isotopes, it is relatively easy to separate the two. There is no such possibility in the



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core of a typical nuclear reactor in which the fissionable material, e.g.,  $U^{235}$ , is a hard oxide or metal enclosed in the cladding of fuel elements. Other radioactive components with half-lives short compared to krypton-89 can also be readily separated.

5                   The high productivity of this method is primarily the result of: (1) the large cross-section of the decay reaction (n, f) of up to  $600-800 \times 10^{-24}$  for thermal neutrons for such nuclei as  $U^{235}$ ,  $U^{233}$ , or  $Pu^{239}$ ; and (2) the ability to remove the krypton-89 from other gaseous end products of the reaction due to differential decay. For a unit target, this method is  
10                   about 1000 times more efficient than the prior art. Because the half-life of krypton-89 (190.7 seconds) is significantly longer than that of krypton-90 (32.2 seconds), it is possible to decrease the content of strontium-90 in the mixture to about  $10^{-4}$  atomic percent, providing for high radioisotope purity in the strontium-89.

15                   The method of strontium extraction via a continuous gas loop is illustrated in FIG. 2. The process is begun after the transitional processes bound up with the reactor start-up are finished (about 20 minutes). Referring to FIG. 2, valves 3 and 9 are opened and a gas pump 5 is turned on. Gas from above the fuel solution is moved to a delaying  
20                   line 4. The delaying line is designed to keep the gas from arriving at the precipitation device 7 for the time necessary for krypton-90 to decay to strontium-90, thereby removing it from the gas mixture. Rubidium and

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strontium isotopes that have not precipitated in the delaying line settle in the filter 6. The diameter of the delaying line pipe is determined by the condition of laminar gas flow in the pipe. The pipe's length is determined by the delay time for a preset gas flow rate. (If the gas flow rate is about 2 l/min, a delay time of ten minutes is achieved when the pipe inner diameter is 10 mm and the pipe length is 255 meters. If the diameter were 20 mm, a delay line length of 64 meter would give a 10-minute delay.) A ten minute delay yields a radionuclide purity ( $\text{Sr}^{90}/\text{Sr}^{89}$ ) of about  $3 \times 10^{-8}$ .

After going through the delaying line, the gas arrives at the strontium-89 precipitation device 7. The precipitation device is another pipe whose diameter and length are designed for a delay period sufficient for the remaining krypton-89 to decay to strontium-89. This would be about 11 minutes at a gas flow rate of 2 l/minute. Those isotopes of rubidium and strontium, which have not precipitated in the precipitation device, pass through it and settle in the filter 8. The gas, less those fission fragments that have precipitated out or otherwise been removed, is return to the reactor. After the cycle of strontium-89 production is completed, the valves 3, 9 are closed. Strontium-89 deposited in the precipitation device and in the filter 8 are subsequently extracted.

The circulating gas flow removes water vapor from the fuel solution. The initial part of the gas pipe 10 shown in FIG. 2 is inclined so that water vapor is condensed on the pipe wall and the water runs back

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into the reactor vessel by gravity preventing fuel solution water loss. A trap **11** is indicated in FIG. 2 at the entrance to the gas loop to hinder non-gaseous fission fragments moved by the gas flow over the fuel solution from getting into the gas loop.

5                                If the precipitation rate of strontium-89 is high, most of it will accumulate in the precipitation device **7**. An acid solution can then be used to wash out strontium-89 from which it is subsequently extracted and subjected to radiochemical purification. If the precipitation rate is low, most of the strontium-89 will accumulate in the filter **8**. This filter can  
10 consist of thin, fine nets of stainless steel. The strontium-89 can then be extracted by pumping an acid solution through the filter. Alternatively, a removable filter could be used with extraction of the strontium-89 being done at a later time.

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CLAIMS:

1. A method of extracting strontium-89 from a uranyl sulfate water solution fueled nuclear reactor, the method characterized by:

5 operating said solution nuclear reactor whereby inert gaseous fission fragments are produced and migrate to the free volume above the solution surface, said gaseous fission fragments comprised of isotopes of beryllium, krypton and rubidium;

10 pumping said inert gaseous fission fragments through a first delaying device at a flow rate sufficiently slow to allow a desired percent of radioactive krypton-90 to decay to strontium-90, whereby said strontium-90 is precipitated out of the gas;

15 passing gas through a first filter to remove rubidium and strontium isotopes that were not precipitated in said first delaying device;

pumping remaining gas through a second delaying device (strontium-89 precipitation device) at a flow rate sufficiently slow to allow the decay of the remaining krypton-89 to strontium-89, whereby the desired strontium-89 is precipitated out;

20 passing gas through a second filter to remove any remaining rubidium and strontium isotopes that were not precipitated out

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in said strontium-89 precipitation device;

pumping remaining gas back to the reactor; and

extracting precipitated strontium-89 from said strontium-89 precipitation device and from said second filter.

5                   2.     The method of claim 1 characterized in that said first delaying device is comprised of a pipe whose inner diameter and length are calculated for a given gas flow rate to contain the gas for a time sufficient to allow the decay of essentially all the krypton-90 to strontium-90.

10                   3.     The method of claim 1 characterized in that said second delaying device is comprised of a pipe whose inner diameter and length are calculated for a given gas flow rate to contain the gas for a time sufficient to allow the decay of a desired percentage of the krypton-89 to strontium-89.

15                   4.     The method of claim 1 characterized in that the extraction of strontium-89 from said strontium-89 precipitation device and said second filter is by an acid wash.

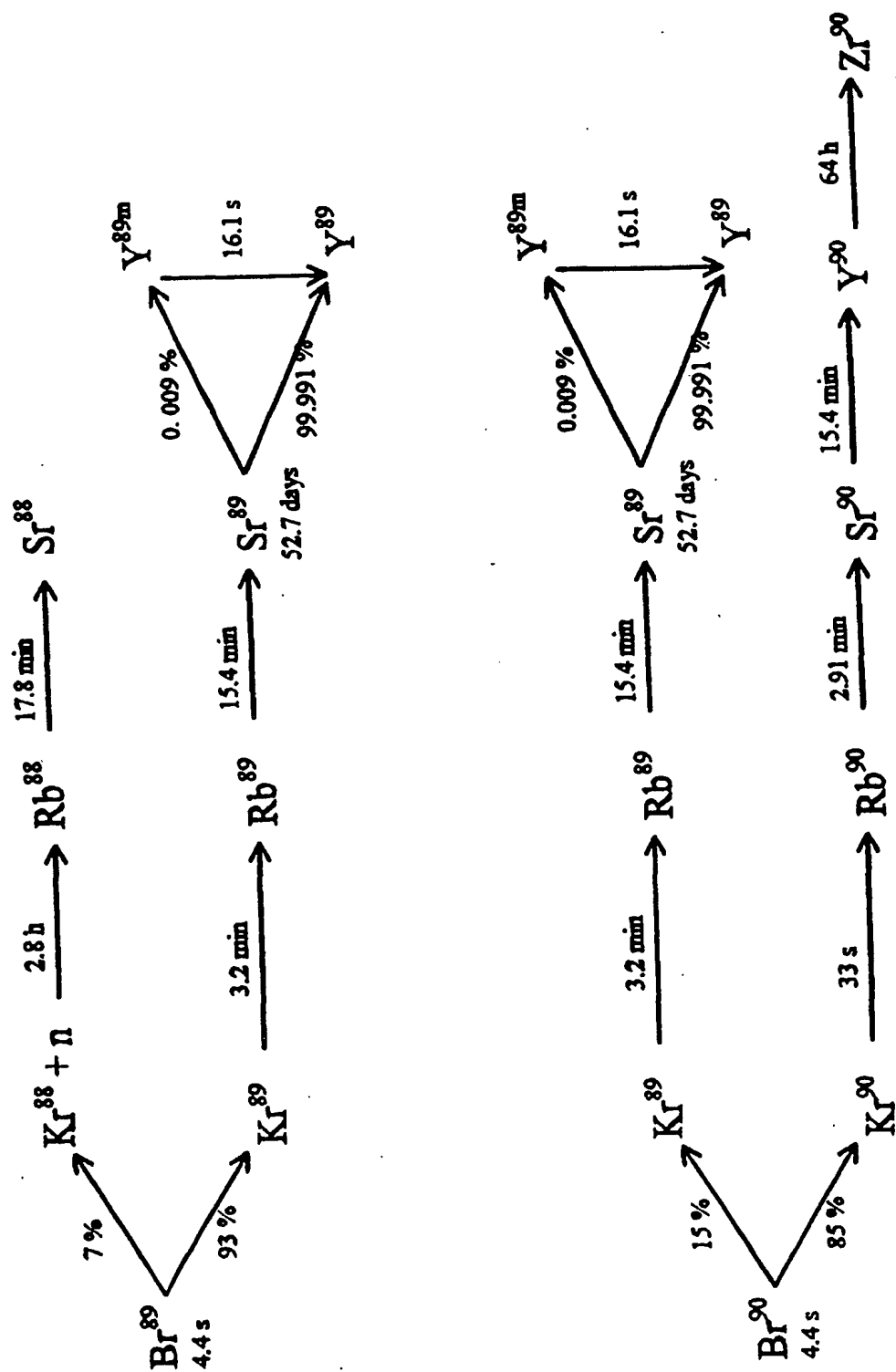


FIG. 1A

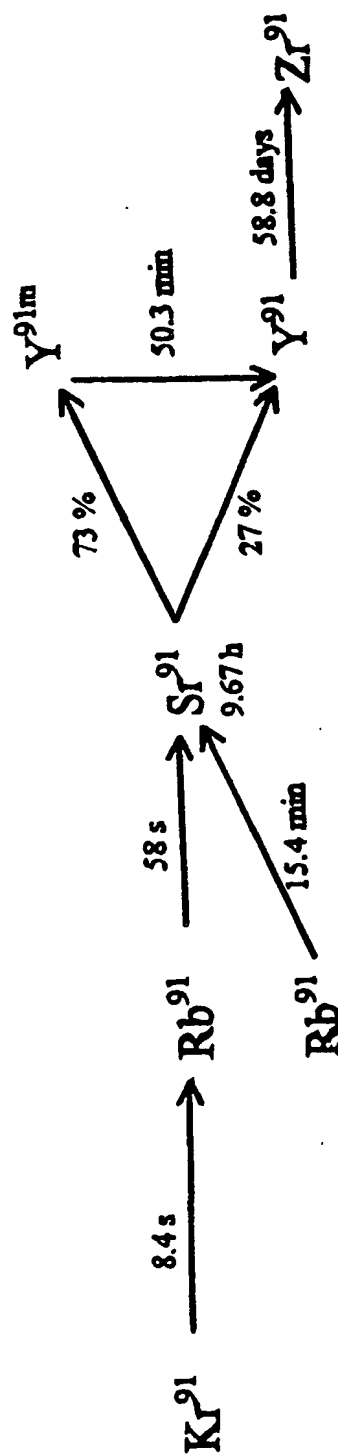


FIG. 1B

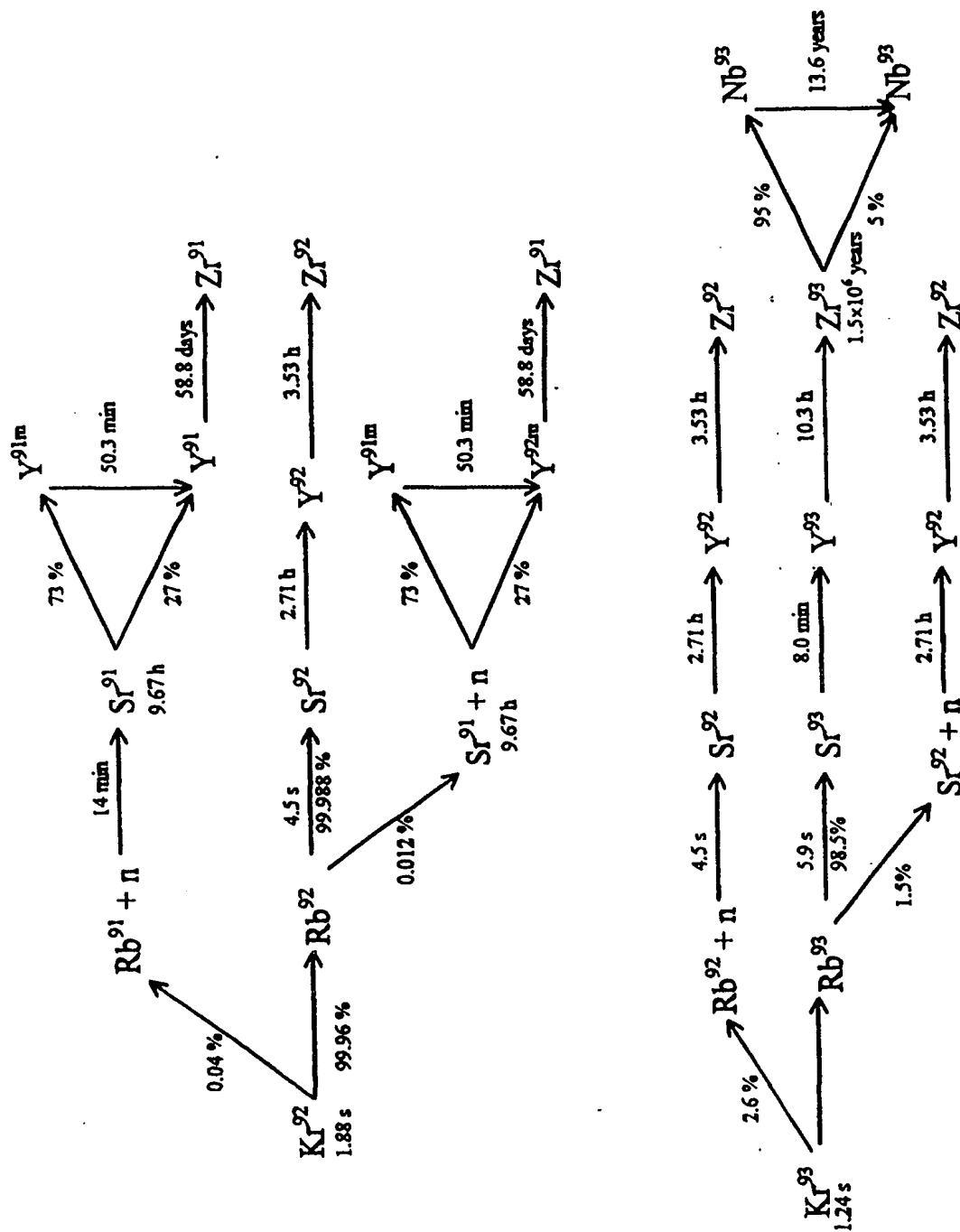


FIG. 1C



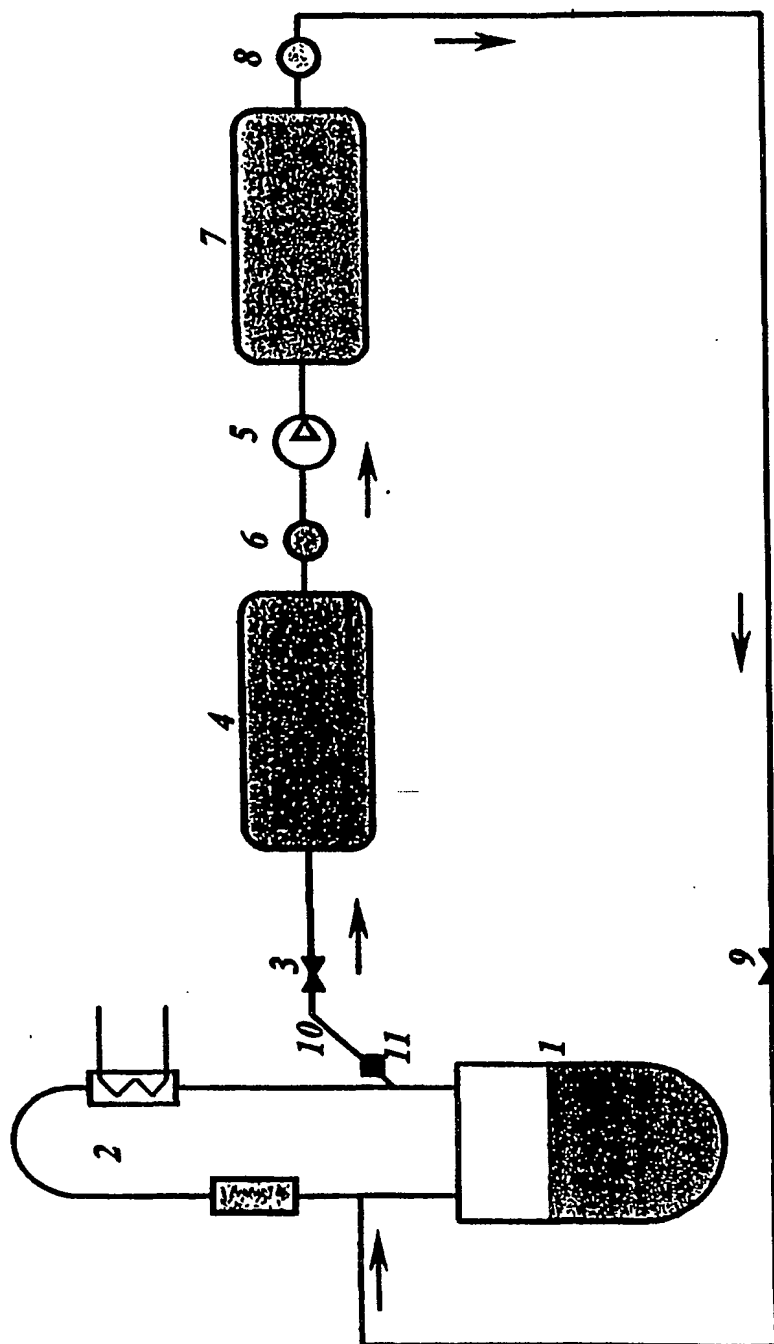


FIG. 2

## INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US00/19574

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) :G21C 19/00

US CL :376/189

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 376/189, 311, 313, 314, 354-358; 423/262

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 3,080,307 A (RINALD) 05 MARCH 1963, SEE ENTIRE DOCUMENT.	1-4
Y	US 3,166,478 A(LINDSTROM) 19 JANUARY 1965, SEE ENTIRE DOCUMENT.	1-4
Y	US 2,860,093 A (WIGNER ET AL) 11 NOVEMBER 1958, SEE ENTIRE DOCUMENT.	1-4
Y	US 2,945,794 A (WINTERS ET AL) 19 JULY 1960, SEE ENTIRE DOCUMENT.	1-4
Y	US 3,791,107 A (GUSTAVSSON) 12 FEBRUARY 1974, SEE ENTIRE DOCUMENT.	1-4
Y	US 3,803,802 A (SCHROTER ET AL) 16 APRIL 1974, SEE ENTIRE DOCUMENT.	1-4

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Date of the actual completion of the international search

29 SEPTEMBER 2000

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## INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US00/19574

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 3,871,841 A (QUEISER ET AL) 18 MARCH 1975, SEE ENTIRE DOCUMENT.	1-4
Y	US 3,074,776 A (RYAN ET AL) 22 JANUARY 1963, SEE ENTIRE DOCUMENT.	1-4
Y	US 3,944,646 A (MARTIN) 16 MARCH 1976, SEE ENTIRE DOCUMENT.	1-4
Y	US 5,875,220 A (ZHUIKOV ET AL) 23 FEBRUARY 1999, SEE ENTIRE DOCUMENT.	1-4
Y	US 5,596,611 A (BALL) 21 JANUARY 1997, SEE ENTIRE DOCUMENT.	1-4
Y	US 3,074,776 A (RYAN ET AL) 22 JANUARY 1963, SEE ENTIRE DOCUMENT.	1-4

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